CLAIMS

What is claimed is:

1. A method for producing single and multi-walled Carbon Nanotubes (CNT)s and composite CNTs from the gas phase comprising
   one or more CNT reactors;
   one or more sources supplying energy to said CNT reactor(s);
   one or more sources of pre-made aerosol catalyst particles introduced to said CNT reactor(s) wherein the catalyst particles are produced by physical vapor nucleation of catalyst material or by solution droplet thermal decomposition of catalyst precursor or are aerosolized from a powder or suspension or wherein the catalyst particles are produced by a chemical method and are subsequently pre-classified according to one or more particle properties;
   one or more carbon sources introduced to said CNT reactor(s);
2. A method of claim (1) additionally including one or more of
   one or more pre-reactors for producing pre-made catalyst particles;
   one or more catalyst particle classifiers;
   one or more CNT samplers;
   one or more CNT classifiers;
   one or more sources supplying energy to said pre-reactor(s);
   one or more reagents supplied to said CNT reactor(s)/pre-reactors(s);
   one or more aerosol samplers and/or classifiers extracting all or part of said CNT aerosol flow;
   one or more additives to said CNT reactor(s) and/or pre-reactors(s) to produce a composite CNT aerosol;
   one or more aerosol samplers and/or classifiers extracting all or part of said composite CNT aerosol flow.
3. A method of claim (1) wherein the catalyst precursor contains one or more metals.
4. A method of claim (1) wherein the catalyst particles are formed due to the nucleation of supersaturated vapor wherein the vapor is evaporation from one or more resistively heated wires consisting of one or more metals or metal alloys, due to metal or alloy laser ablation, due to metal or alloy arc, spark or electrostatic discharge, due to evaporation from a conductively heated metal or alloy or due to evaporation from radiatively heated metal or alloy.

5. A method of claim (4) wherein the supersaturation is created by means of gas cooling by convective, conductive and/or radiative heat transfer and/or adiabatic expansion.
6. A method of claim (3) wherein the catalyst precursor is a metalorganic, organometallic or inorganic catalyst containing compound.
7. A method of claims (1) or (2) wherein the pre-made catalyst particles are classified according to one or more particle properties.
8. A method of claim (7) wherein the pre-made catalyst particles are mobility-size classified, mass classified, solubility classified, reactivity classified, inertially classified, thermophoretically classified, diffusionally classified, charge classified, crystalinity classified and/or gravitationally classified.
9. A method of claim (8) wherein the pre-made catalyst particles are classified by a differential mobility analyzer or by a mass spectrometer.
10. A method of claims (1) or (2) wherein the carbon source is an organic or inorganic carbon containing compound.
11. A method of claim (10) wherein the organic compound is a hydrocarbon.
12. A method of claim (11) wherein the hydrocarbon is methane, ethane, propane, acetylene, ethylene, benzene, toluene, o-xylene, p-xylene, 1,2,4-trimethylbenzene, 1,2,3-trimethylbenzene, $C_{15}H_{32}$, $C_{16}H_{34}$, $C_{17}H_{36}$, or $C_{18}H_{38}$.
13. A method of claim (10) wherein the organic compound is an oxygen containing compound.
14. A method of claim (13) wherein the oxygen containing compound is methanol, ethanol, propanol, butanol, pentanol, hexanol, heptanol, octanol, acetone, methyl ethyl ketone, formic acid or acetic acid.
15. A method of claim (10) wherein the inorganic compound is carbon monoxide CO.
16. A method according to claims (1) or (2) wherein the residence time and/or temperature and/or catalyst particle properties and/or catalyst particle concentration and/or reagent concentration and/or carbon source concentration histories in one or more CNT reactors are controlled and the pre-made catalyst particles, carbon sources, reagents and carrier gases are continuously introduced into the CNT reactor which is maintained at steady state conditions and the products are continuously evacuated from the CNT reactor(s) and or pre-reactor(s) to comprise a continuous production of product or the pre-made catalyst particles, carbon sources, reagents and carrier gases are periodically introduced into the CNT reactor in which the conditions are controlled for a period of time and the

products are periodically evacuated from the CNT reactor(s) and or pre-reactor(s) to comprise a batch production of product.

17. A method according to claim (16) wherein the reactor length, volume and/or wall temperature and/or the flow rate of carbon sources and/or reagents and/or carrier gases are used to control the residence time and/or temperature history of catalyst particles and/or CNTs and/or composite CNTs in the CNT reactor(s) and or pre-reactor(s).
18. A method of claims (17) wherein said CNT reactor(s) and/or pre-reactor(s) use sheath gas introduced through a porous or perforated wall, a co-flowing channel or an injection port to control the aerosol flow so as to minimize deposition and/or to control the residence time, gaseous environment and/or temperature history of catalyst particles and/or CNTs and/or carbon nanotube composites inside the CNT reactor(s) and or pre-reactor(s).
19. A method of claim (2) wherein said CNT aerosol sampler and/or composite CNT sampler selectively extracts a portion of carbon nanotubes and/or carbon nanotube composites from inside the CNT reactor(s).
20. A method of claim (19) wherein said sampling is in the form of one or more isokinetic sampling probes or one or more sampling probes combined with one or more particle aerodynamic lenses and/or one or more particle acoustic lenses.
21. A method of claims (1) or (2) wherein the CNT reactor and/or pre-reactor surfaces contain material included in one or more catalyst particles or where the CNT reactor and/or pre-reactor surfaces are saturated with material included in one or more catalyst particles.
22. A method of claim (2) wherein the reagent(s) is/are used for participation in a chemical reaction with one or more catalyst particle precursors and/or with one or more pre-made particles and/or with one or more carbon source and/or with amorphous carbon deposited on CNTs and/or with CNTs.
23. A method of claim (22) wherein the chemical reaction of the reagent(s) with catalyst particle precursor and/or with pre-made particles is/are used for promotion of CNT formation and/or where the chemical reaction of the reagent(s) with amorphous carbon is/are used for CNT purification and/or where the chemical reaction of the reagent(s) with the CNTs is/are used for CNT functionalization and/or CNT doping.
24. A method of claim (22) wherein one or more reagents act also as a carbon source.

25. A method of claims (23) and/or (24) wherein the reagent is an alcohol, H_2, H_2O, NO, CO_2, PH_3 and/or NH_3.
26. A method of claims (1) or (2) wherein the energy source is laser, electrical, resistive, conductive, radiative (in the entire range of the electromagnetic spectrum) and/or acoustic heating, combustion or chemical reaction, or nuclear reaction.
27. A method of any of claims (1) to (26) wherein the carrier gas and reagent gases entering the pre-reactor(s) are nitrogen and hydrogen and where the volume percent of hydrogen is preferably between 0.1% and 25% and more preferably between 1% and 15% and more preferably between 5% and 10%and most preferably approximately 7% and where there is one pre-reactor operated in series with one CNT reactor that is aligned with gravity and where the pre-reactor uses a hot wire generator to produce pre-made catalyst particles and where the hot wire generator has a wire diameter between 0.01 and 10 mm and more preferably between 0.2 and 0.5 mm and more preferably approximately 0.25 mm and where in the CNT reactor is essentially circular in cross section, oriented approximately vertically with respect to gravity and has an inner diameter preferably between 0.5 and 50 cm and more preferably between 1.5 and 3 cm and most preferably approximately 2.2 cm and a length preferably between 5 and 500 cm and more preferably between 25 and 200 cm and most preferably approximately 90 cm and where the CNT reactor wall is heated resistively.
28. A method of claim (27) wherein the hot wire generator is separated in space from the CNT reactor and where in the carbon source is CO and where the CO is introduced into the CNT reactor at a normalized volume flow rate of preferably between 5 and 5000 cm^3/min and more preferably between 250 and 800 cm^3/min and most preferably at approximately 400 cm^3/min and where the maximum CNT reactor wall temperature is between 600 and 15000 degrees C and more preferably between 850 and 5000 degrees C and most preferably at approximately 1200 degrees C and where in the flow rate through the pre-reactor is between 5 and 5000 cm^3/min and more preferably between 250 and 600 cm^3/min and most preferably at approximately 400 cm^3/min and where the secondary and tertiary reagents are thiophene and octanol and where the thiophene vapor pressure is most preferably between 1 and 1000 Pa and more preferably between 10 and 100 Pa and more preferably between 20 and 40 Pa and most preferably approximately 30 Pa and where the octanol vapor pressure is most preferably between 0.1 and 100 Pa and more preferably

between 1 and 10 Pa and more preferably between 2 and 4 Pa and most preferably approximately 3.4 Pa.

29. A method of claim (27) wherein the pre-reactor is essentially circular in cross section, is smoothly integrated with the CNT reactor by inserted it therein and aligning said pre-reactor with the centerline of said CNT reactor and where the hot wire generator is located essentially at the exit of the pre-reactor and where the end of the smoothly integrated pre-reactor is preferably located where the CNT reactor wall temperature is between 0 and 5000 degrees C and more preferably between 350 and 450 degrees C and most preferably approximately 400 degrees C and where the inner diameter of the pre-reactor is preferrably between 0.1 and 5 cm and more preferably between 0.5 and 1.5 cm and most preferably approximately 0.9 cm and where the outer diameter of the pre-reactor is preferably between 0.2 and 10 cm and more preferably between 0.5 and 2.0 cm and most preferably approximately 1.3 cm and where the maximum CNT reactor wall temperature is between 600 and 15000 degrees C and more preferably between 850 and 1500 degrees C.
30. A method of claim (29) where in the carbon source is CO and wherein the CO is introduced into the CNT reactor around the pre-reactor at a normalized volume flow rate of preferably between 5 and 5000 cm^3/min and more preferably between 250 and 800 cm^3/min.
31. A method of claim (29) wherein the inner flow rate through the pre-reactor is between 5 and 5000 cm^3/min and more preferably between 250 and 600 cm^3/min and most preferably at approximately 400 cm^3/min.
32. A method of claims (21) and (31) wherein the CNT reactor walls are constructed from stainless steel.
33. A method of claim (29) wherein the carbon source and a second reagent is ethanol and wherein the ethanol vapor pressure is preferably between 1 and 10000 Pa and more preferably between 100 and 500 Pa and most preferably between 150 and 300 Pa and most preferably approximately 213 Pa.
34. A method of claim (29) wherein the carbon sources and secondary and tertiary reagents are ethanol and thiophene and where the thiophene vapor pressure is most preferably between 0.01 and 1000 Pa and more preferably between 0.1 and 30 Pa and more preferably between 0.2 and 15 Pa and where the ethanol vapor is pressure most preferably between 1 and 20000 Pa and more preferably between 10 and 10000 Pa and more preferably between 50 and 5000 Pa.

35. A method of claims (23) and (32) wherein the reagent for the promotion of CNT growth and functionalization is hydrogen and wherein the volume percent of hydrogen in the hotwire generator is greater than 50% and more preferably greater than 90 % and more preferably greater than 99%.
36. A method of claims (23) and (32) wherein the reagent for the functionalization of carbon nanotubes is water vapor, wherein the water vapor is introduced in the outer CO flow via a saturator and where the concentration of water vapor is between 1 and 10000 ppm and more preferably between 10 ppm and 1000 ppm and more preferably between 100 and 200 ppm and most preferably approximately 150 ppm.
37. A method of any of claims (1) to (37) wherein there are two or more existing pre-made catalyst particle supplies which are composed of particles of essentially similar sizes, compositions, concentrations, states and/or morphologies or are composed of two or more distinct sizes, compositions, concentrations, states and/or morphologies.
38. A method of any of claims (2) to (37) wherein there are two or more pre-reactors and said pre-reactors are operated in parallel and said parallel pre-reactors are operated at essentially similar conditions and/or with essentially similar materials so as to produce pre-made catalyst particles of essentially similar sizes, compositions, concentrations, states and/or morphologies or said parallel pre-reactors are operated at different conditions and/or with different materials and/or methods so as to produce pre-made catalyst particles of two or more distinct sizes, compositions, concentrations, states and/or morphologies.
39. A method of any of claims (1) to (37) wherein said CNT reactors are operated in parallel and said parallel reactors are operated at essentially similar conditions and/or with essentially similar materials so as to produce CNTs with essentially similar length, diameter, morphology and/or chirality or said parallel reactors are operated at different conditions and/or with different materials and/or methods so as to produce CNTs with two or more distinct lengths, diameters, morphologies and/or chiralities.
40. Carbon nanotubes prepared according to any of claims (1) to (36).
41. Carbon nanotubes of claim (37) wherein the length, diameter, number of walls, chirality, purity, and/or composition of dopants and/or attached functional groups are controlled.
42. Functionalized carbon nanotubes according to claim (37) wherein the attached functional groups are fullerenes, CNTs, transition metals, transmission metal oxides, polymers and/or polymer catalysts.

43. Carbon nanotubes according to claim (41) where in the geometric standard deviation of the length is less than 2.5 or more preferably less than 1.5 or most preferably less than approximately 1.25 and where in the geometric standard deviation of the diameter is less than 2.5 or more preferably less than 1.75 or most preferably less than approximately 1.4 and where the geometric mean diameter is preferably between 0.4 and 25 nm and more preferably between 0.75 and 5 nm and most preferably between approximately 0.8 and 1.3 nm and where the geometric mean length is preferably between 2 nm and 1 m and more preferably between 10 nm and 1000 nm and more preferably between 25 nm and 100 nm and most preferably between approximately 45 and approximately 55 nm.
44. Carbon nanotubes of claims (37) through (43), wherein the carbon nanotubes are coated with one or more additive solids or liquids and/or solid or liquid particles to constitute a carbon nanotube composite.
45. CNT composites of claim (44) wherein one or more additive is introduced to the CNT reactor in the gas phase as a gas and/or as a liquid or solid aerosol particle and/or wherein one or more additive gases are supersaturated so as to condense onto the CNT and/or wherein one or more additive gases chemically react with the surface of the CNT and/or with another additive, and/or with a functional group and/or with a doping material of the CNT and/or wherein one or more additive aerosol particles are attached to the surface of the CNT to form a liquid, solid or mixed coated CNT or a CNT-additive particle agglomerate or a mixture thereof.
46. Carbon nanotube composites of claim (45) wherein the coating material is a metal, a polymer, an organic, a ceramic or a mixture thereof.
47. Carbon nanotubes and/or carbon nanotube composites of any of claims (37) to (46), wherein the carbon nanotubes and/or composite carbon nanotubes are formulated as a dispersion in a gas, a dispersion in a liquid, a dispersion in a solid, a powder, a paste or a colloidal suspension or are deposited on a surface.
48. A functional material made with a formulation of claim (47).
49. A thick or thin film, a line, a wire or a layered structure composed of a functional material of claim (48)
50. A thin or thick film, a line, a wire or a structure of claim (49) deposited by electrical, acoustic, thermophoretic, inertial, diffusional, turbophoretic and/or gravitational forces.
51. A thin or thick film, a line, a wire or a structure of claim (50) wherein the deposition is enhanced by jet focusing.

52. A thin or thick film, a line, a wire or a structure of any of claims (44) to (51) wherein the coating material is composed of one or more monomers and zero or more catalysts and the resulting functional material is heated so as to induce polymerization.
53. A device made according to any of claims (37) to (52).
54. A device of claim (53) wherein the device is an electrode of a fuel cell or battery, a heat sink or heat spreader, a metal-matrix composite or polymer-matrix composite in a printed circuit or electron emitter in a field emission display.